

Nuclear Engineering and Technology

journal homepage: www.elsevier.com/locate/net

Original Article

Characteristics of Plastic Scintillators Fabricated by a Polymerization Reaction



Cheol Ho Lee, Jaebum Son, Tae-Hoon Kim, and Yong Kyun Kim*

Department of Nuclear Engineering, Hanyang University, 222 Wangsimni-ro, Seongdong-gu, Seoul 04763, South Korea

ARTICLE INFO

Article history:

Received 27 April 2016

Received in revised form

9 September 2016

Accepted 3 October 2016

Available online 13 October 2016

Keywords:

Compton Edge

Emission Wavelength

Light Output

Plastic Scintillator

Polymerization

ABSTRACT

Three plastic scintillators of 4.5 cm diameter and 2.5-cm length were fabricated for comparison with commercial plastic scintillators using polymerization of the styrene monomer 2,5-diphenyloxazole (PPO) and 1,4-bis benzene (POPOP). Their maximum emission wavelengths were determined at 426.06 nm, 426.06 nm, and 425.00 nm with a standard error of 0.2% using a Varian spectrophotometer (Agilent, Santa Clara, CA, USA). Compton edge spectra were measured using three gamma ray sources [i.e., cesium 137 (^{137}Cs), sodium 22 (^{22}Na), and cobalt 60 (^{60}Co)]. Energy was calibrated by analyzing the Compton edge spectra. The fabricated scintillators possessed more than 99.7% energy linearity. Light output was comparable to that of the BC-408 scintillator (Saint-Gobain, Paris, France). The fabricated scintillators showed a light output of approximately 59–64% of that of the BC-408 scintillator.

Copyright © 2016, Published by Elsevier Korea LLC on behalf of Korean Nuclear Society. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

1. Introduction

A wide range of scintillation materials are used in various fields of medicine and security and for scientific purposes in research institutions. Examples of such purposes are medical imaging, ionizing radiation detection, and spectroscopy. Scintillators can be composed of organic or inorganic materials in combination with solvents. Gaseous materials can also be used for scintillation counting [1]; the most common example is helium 3 (^3He) counters used for neutron detection [2]. Scintillation materials are typically liquid, plastic, or crystal. Plastic scintillators are more durable than liquid scintillators and can be machined into nearly any shape. They

have many advantages such as fast rise and decay times, high optical transmission, ease of manufacturing, low cost, and large available size. Because of these characteristics, there has been an increased interest in developing plastic scintillators and an interest in their many applications in nuclear physics and radiation detection, and particle identification [3]. The most common preparation method for plastic scintillators is thermal polymerization of a solution containing a liquid monomer. The polymerization techniques vary with the composition and size of the desired sample. The polymerization is initiated slowly at a low temperature and then completed at a high temperature. In this study, three plastic scintillators 4.5 cm in diameter and 2.5 cm in length were

* Corresponding author.

E-mail address: ykkim4@hanyang.ac.kr (Y.K. Kim).

<http://dx.doi.org/10.1016/j.net.2016.10.001>

1738-5733/Copyright © 2016, Published by Elsevier Korea LLC on behalf of Korean Nuclear Society. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

fabricated by the polymerization of the styrene monomer 2,5-diphenyloxazole (PPO) and 1,4-bis benzene (POPOP). Gamma ray spectra were measured using standard gamma ray sources such as cesium 137 (^{137}Cs), sodium 22 (^{22}Na), and cobalt 60 (^{60}Co). Energy was calibrated by analyzing the pulse spectra. The purpose of the energy calibration was to convert the channels in the pulse spectra into gamma ray energy. Relative light output was estimated to compare the fabricated scintillators with a commercial scintillator (BC-408 scintillator; Saint-Gobain, Paris, France).

2. Materials and methods

2.1. The plastic scintillator preparation process

Three plastic scintillators were fabricated through polymerization to compare properties such as emission wavelength and scintillation efficiency with those of commercial plastic scintillators. The recipe used in this study requires three components. The first component is a liquid monomer, which is the transparent liquid. A commercially available styrene monomer with 99.5% purity was the solvent. The second component is commercially sold as 2,5-diphenyloxazole (i.e., PPO) in the form of a white powder and is a scintillating chemical whose peak emission wavelength is 303 nm, which lies within the UV spectrum. The third component is POPOP [i.e., 1,4-bis[2-(phenyloxazoly)]-benzene] which is a light yellow secondary scintillating material. The POPOP component acts as a scintillator and as a wavelength shifter, which means that it converts the shorter wavelengths emitted from the PPO into longer wavelengths. Its wavelength peak is at 410 nm, which is a visible violet light. The styrene monomer was mixed with PPO and POPOP. Table 1 shows the masses of the components used for the preparation of plastic scintillators, as measured by an electronic scale. With a density of 0.906 g/mL, 100 g of styrene are equivalent to 110.375 mL. Approximately 80 mL of the styrene monomer are needed to create a plastic scintillator of 2.5 cm in length. The reference denotes the masses of the additives (i.e., PPO and POPOP) for 80 mL of styrene. The mixed solution was poured into 100-mL beakers to create plastic scintillators of 4.5 cm diameter and 2.5 cm length, as shown Fig. 1A. The solution was stirred with a stirrer for 6 hours, and was then stirred inside a 60°C water bath. The solution was afterwards placed in a high temperature heater to induce the polymerization reaction. For

complete dissolution, the temperature of the heater had to be maintained at 100°C for 2 hours because stirring alone was insufficient. After this procedure, the temperature of the heater was increased to 120°C for 150 hours while polymerization occurred. After the polymerization reaction had ended, a cooling process was allowed for 60 hours inside the heater. The temperature of the heater was gradually decreased to prevent generating air bubbles caused by internal stress inside the polystyrene material. Fig. 2 shows the temperature profile of the heater. A plastic scintillator that was fabricated using this method is in Fig. 1B. The plastic scintillator was cut by a cutting machine (Minisaw; GLP Korea, Gwangmyeong, Korea) to remove air bubbles generated on the top and bottom surfaces. The surfaces were then polished with 800–4000 grit sandpaper using a high-speed rotating machine (twin variable speed grinder-polisher; Buehler, Lake Bluff, IL, USA). Figs. 1C and 1D show the plastic scintillator after polishing and the scintillator wrapped in Teflon tape as a reflector for protection, respectively.

2.2. Experimental setup

Various experiments were conducted to evaluate the key characteristics of fabricated plastic scintillators such as emission wavelength, linearity, and light output. First, a fluorescence spectrophotometer manufactured by Varian Cary Eclipse (Agilent, Santa Clara, CA, USA) was employed to measure the emission wavelength of the scintillator. After being placed inside the spectrophotometer, the scintillator was irradiated with an excitation beam to induce it to emit light, the intensity of which was recorded as a graph by the Cary Eclipse software (Agilent). Second, three gamma ray sources (^{137}Cs , ^{22}Na , and ^{60}Co) were used to evaluate the linearity of the scintillators through energy calibration. The purpose of the energy calibration was to convert the channels in the pulse spectra measured by the multichannel analyzer (MCA) module into gamma ray energy; the linearity was subsequently estimated, based on the calibration. Third, the relative light output of the scintillator was calculated by using the Bertolaccini method [4, 5]. Four parameters are necessary to use the method. Section 3.3 provides a detailed introduction.

3. Results and discussion

3.1. Emission wavelength

In general, typical commercial plastic scintillators have a peak emission at a wavelength of 425 nm [6, 7]. Fig. 3 shows the emission wavelength intensities of the three plastic scintillators measured with the fluorescence spectrophotometer. The beam wavelength of the spectrophotometer was set to 310 nm (i.e., the plastic scintillator absorption wavelength) to the emission wavelength spectra, and the beam was aimed at the plastic scintillator. Absorption of the beam inside the scintillator induced photoluminescence and this photoluminescence was recorded by the spectrophotometer. The results are presented for plastic scintillators #1–#3 at the peak emission wavelengths of 426.06 nm, 426.06 nm, and

Table 1 – The Masses of the Ingredients Used for the Preparation of the Three Plastic Scintillators.

	Styrene (mL)	PPO (g)	POPOP (g)
Mass ratio	100 g (110.375 mL)	1 g	0.05 g
Reference ^a	80.00	0.727	0.0364
#1 beaker	79.88	0.728	0.0367
#2 beaker	80.24	0.726	0.0370
#3 beaker	80.44	0.727	0.0390
POPOP, 1,4-bis benzene; PPO, 2,5-diphenyloxazole.			
^a The reference denotes the mass of the additives (i.e., PPO and POPOP) for 80 mL of styrene.			

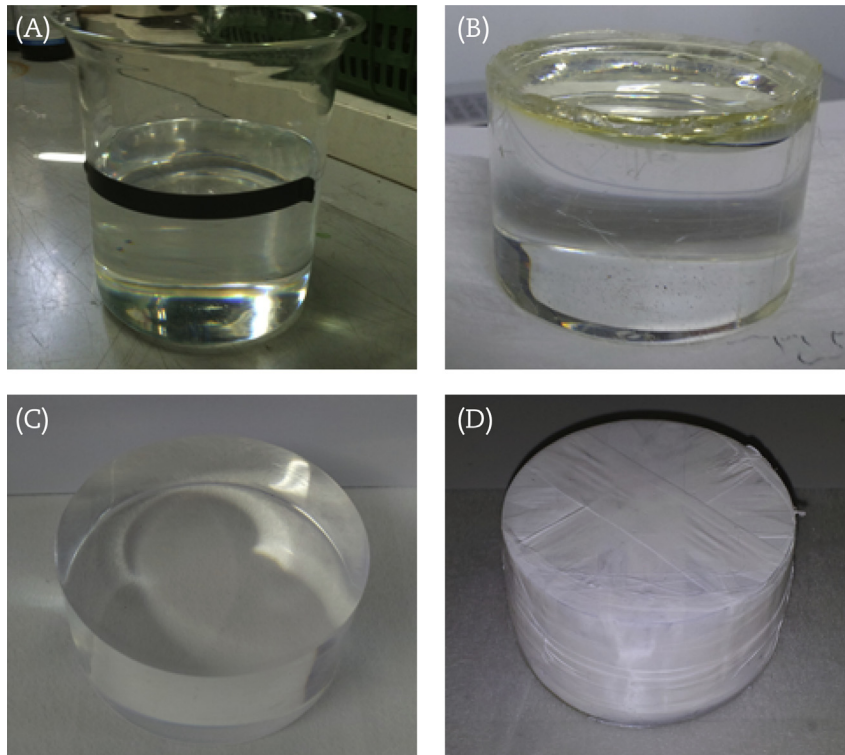


Fig. 1 – The plastic scintillator preparation process. Step-by-step images show (A) the status after stirring, (B) the scintillator completed by polymerization reaction, and (C) the finished plastic scintillator. (D) is the scintillator wrapped in Teflon tape as a reflector for protection.

425.00 nm, respectively, with 0.2% standard error. When measuring the emission wavelength of the plastic scintillator with the spectrophotometer, it is impossible to fully control the angle of the incident beam with respect to the scintillator's surface. This factor may explain the slight difference in intensity. In addition, an intensity difference can occur because the solution was not prepared according to the correct mass ratio (100:1:0.05). However, the fabricated plastic scintillators have the same properties as commercial

scintillators [e.g., BC-408 scintillator (Saint-Gobain)] because they share the same peak emission wavelength.

3.2. Energy calibration of the scintillator detector

A standard gamma source such as ^{137}Cs , ^{22}Na , and ^{60}Co is usually used for the energy calibration of a general radiation

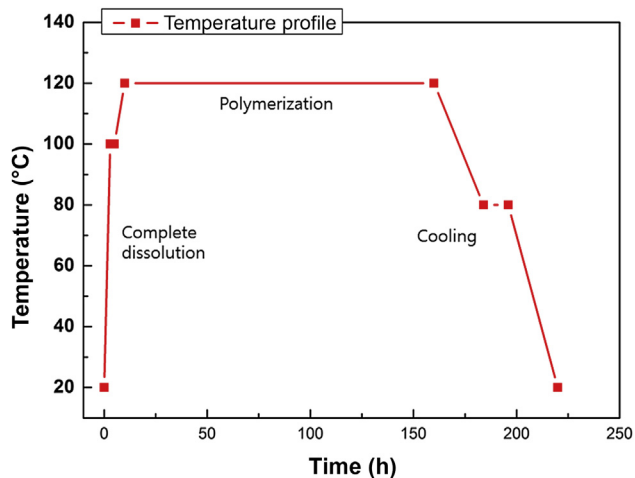


Fig. 2 – Temperature profile of the high temperature heater over the course of the preparation process.

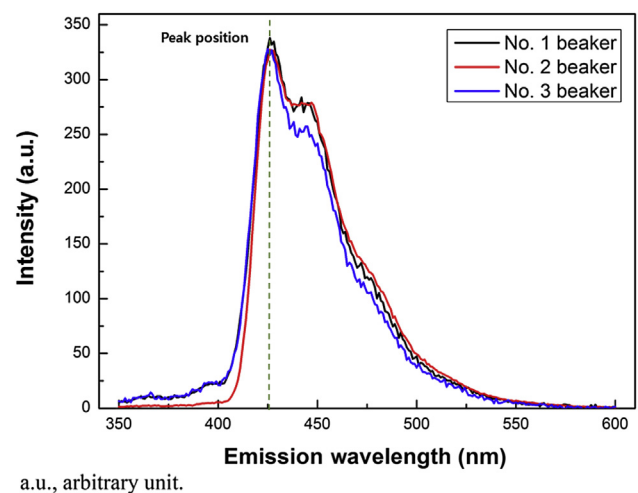


Fig. 3 – Emission wavelength intensities of the three plastic scintillators. Each scintillator shows a peak at a wavelength of approximately 425 nm.

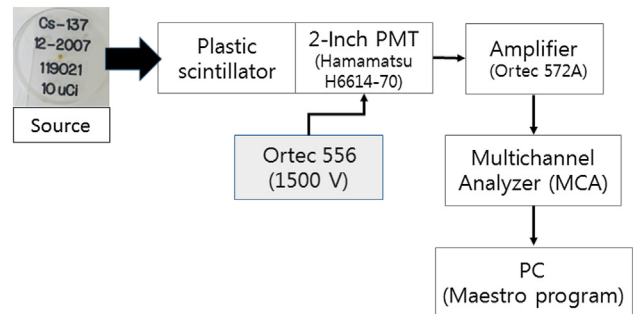
detector. Measurements of the Compton edge spectra using gamma sources were possible in this study because a plastic scintillator does not exhibit phosphorescence. In Compton scattering, a gamma ray is scattered by a free electron. The energy of the electron is transferred to the detector while the scattered gamma ray escapes from the detector. The recoil electron has a well-defined maximum energy—the aforementioned Compton edge, which corresponds to the maximum transferred energy from the gamma ray to the electron. This edge can be used for the energy calibration of the detector and for comparing the relative light output from each scintillator. The Compton edge corresponds to the maximum energy transfer from the scattered gamma ray to the electron. Therefore, the Compton edges of backscattered gamma rays can be calculated by using the kinematic equation of energy conservation and by taking into account that the maximum amount of energy is transferred during the backscattering of the gamma rays. Table 2 shows the calculated Compton edge energy values of the gamma ray sources.

In this work, three radioactive sources were used for energy calibration: ^{22}Na , ^{60}Co , and ^{137}Cs . The Compton edge energy for each source was calculated, based on the gamma ray energies taken from Table of Isotopes [8]. For the ^{60}Co source, an average energy was selected for the Compton edge calculations because it emits two gamma rays of 1.17 MeV and 1.33 MeV. Fig. 4 shows a schematic of the experimental setup for measuring the Compton edge spectra. The specifications of the modules used to measure the Compton edge spectra are as follows. (1) Photonmultiplier tube (PMT): Hamamatsu H6614-70 (Hamamatsu City, Japan; operating voltage, 1500 V). (2) Amplifier: Ortec 572A (Ortec, Inc., Tennessee, OR, USA; shaping time, 0.5 μs). (3) High voltage power supply: Ortec 556 (Ortec, Inc.). (4) Multichannel analyzer: Ortec 919E (Ortec, Inc.).

Fig. 5 shows set up of the modules, based on Fig. 4. The plastic scintillator was attached to the window of the PMT using optical grease. Lead bricks (5 cm thick) provided radiation shielding and black tape blocked external light. Maestro software (Ortec, Inc., Tennessee, OR, USA), which was applied to the MCA, was used to record the spectra over a measurement time of 600 seconds. Fig. 6 shows the gamma ray pulse spectra of ^{137}Cs , ^{22}Na , and ^{60}Co obtained with the three plastic scintillators. To identify the channel, which corresponds to the Compton edge energy in these spectra, the middle point where the slope drops to one-half of the value of the Compton peak was selected. The energy calibration results of each scintillator are as follows: (1) Scintillator #1 \rightarrow Channel = $-32.29554 + 0.79962 \times \text{energy}$ ($R^2 = 99.944$); (2) Scintillator #2 \rightarrow Channel = $-16.22904 + 0.70749 \times \text{energy}$ ($R^2 = 99.951$); and (3) Scintillator #3 \rightarrow Channel = $-41.55461 + 0.80273 \times \text{energy}$ ($R^2 = 99.786$).

Table 2 – The Calculated Compton Edge Values of the Three Gamma Ray Sources.

Source	Activity (μCi)	Gamma energy (keV)	Compton edge energy (keV)
^{22}Na	1.231	511.003	340.110
^{60}Co	3.314	1173.228/1332.492	Avg. 1040.79
^{137}Cs	8.071	661.657	477.334
^{60}Co , cobalt 60; ^{137}Cs , cesium 137; ^{22}Na , sodium 22.			



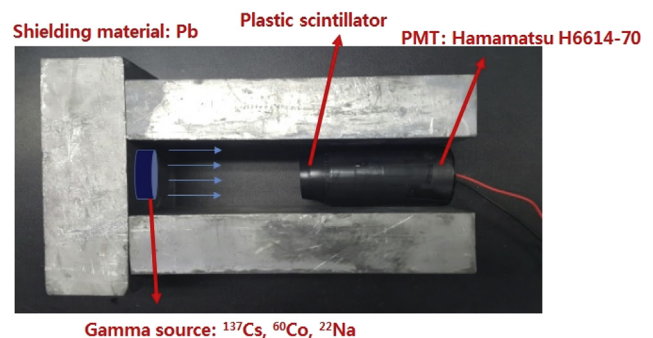
PC, personal computer; PMT, photonmultiplier tube.

Fig. 4 – Schematic of the experimental setup for measuring the Compton edge spectra using the fabricated plastic scintillators.

The energy linearity of each scintillator, obtained through the energy calibration, was determined as 99.944, 99.951, and 99.786; thus, confirming that the fabricated plastic scintillators possess excellent linearity. Because of the energy calibration, the pulse spectra for ^{137}Cs , ^{22}Na , and ^{60}Co can be shown at their correct energy scales (Figs. 6 and 7). The x axes of the pulse spectra that were measured by the MCA module for the energy calibration were converted into gamma ray energy values. The result was in agreement with the general observation that in the low energy region the response of scintillators is proportional to the incoming energy.

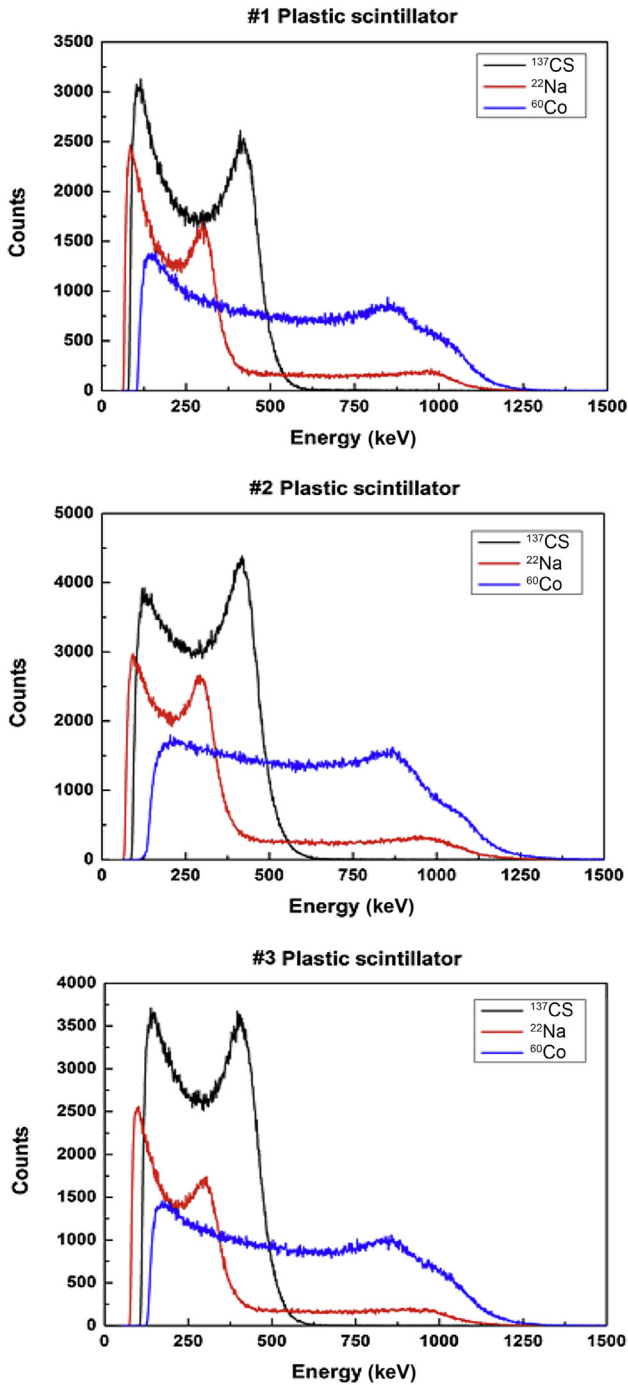
3.3. Light output

The light yield was determined by measuring the number of photoelectrons per energy unit (N_{phe}). This can be achieved through a comparison of the peak position of a single photoelectron spectrum (PP_{phe}) with a characteristic point of any energy spectrum [4, 5]. In the current measurements, the Compton edge (477.334 keV, ^{137}Cs) for the 661.657 keV full energy peak of the ^{137}Cs gamma source was used. Peak positions were recorded with different gains of the spectroscopy amplifier K because of the large amplitude differences between single photoelectron signals and the ^{137}Cs energy spectrum. The number of photoelectrons per energy unit is given by the following equation [4, 5]:



Co, cobalt; Cs, cesium, Na, sodium; Pb, lead.

Fig. 5 – Experimental setup, based on Fig. 4. The photonmultiplier tube (PMT) is connected to a high voltage power supply module (Ortec 556). The distance between the scintillator and source is 5 cm.



Co, cobalt; Cs, cesium; Na, sodium.

Fig. 6 – The gamma ray pulse spectra for three different sources obtained with each fabricated plastic scintillator.

$$N_{phe} = (PP_E/K_E)/(PP_{1phe}/K_{1phe})/0.477334 \text{ (phe/MeV)} \quad (1)$$

in which PP_E is the Compton edge peak position of the ^{137}Cs gamma source, K_E is gain of the spectroscopy amplifier in the experiment. Table 3 shows the light output measured by a commercial plastic scintillator (BC-408, Saint-Gobain) and by the fabricated plastic scintillators. Parameters such as amplifier gain and peak position were obtained through pulse spectra analysis. The quantum efficiency of the PMT should be

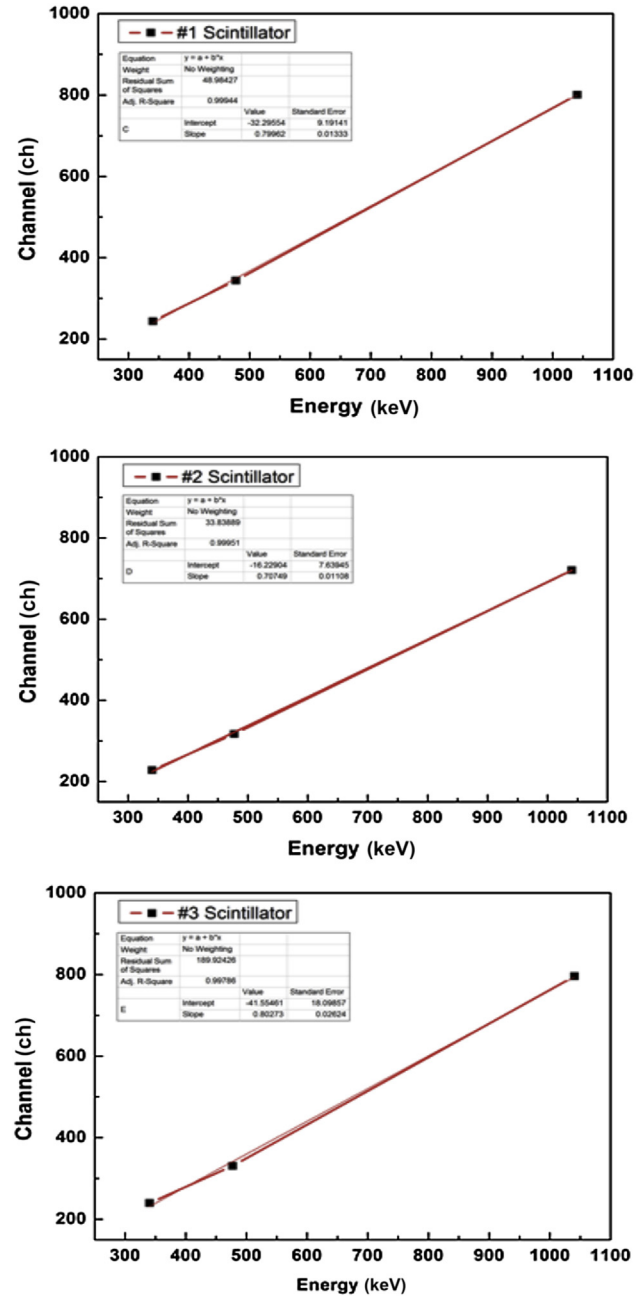


Fig. 7 – Energy calibration via linear fit for the data points corresponding to the Compton edges in Fig. 6. The linearity of scintillators #1–#3 is 99.944, 99.951, and 99.786, respectively.

considered when calculating the light output because there is a difference in the quantum efficiency for different wavelengths. The quantum efficiency is the number of photoelectrons emitted from the photocathode divided by the number of incident photons. The quantum efficiency of the PMT is given by the following equation [9]:

$$QE = (S \times 1240)/\lambda \times 100 \quad (2)$$

in which QE is the quantum efficiency, S is the radiant sensitivity in amperes per watt at the given wavelength,

Table 3 – Light Output Measured with the BC-408 Scintillator and the Fabricated Plastic Scintillators.

Parameter	#1 Scintillator	#2 Scintillator	#3 Scintillator	BC-408
Gain (K_E)	10	10	10	10
Gain ($K_{1\text{phe}}$)	1000	1000	1000	1000
Peak position (PP_e)	344	317	331	644
Peak position ($PP_{1\text{phe}}$)	119	108	121	140
Light yield (phe/MeV)	605.60	614.91	573.09	963.69
Quantum efficiency (%)	20.95	20.95	21.01	21.01
Light output (ph/MeV) ^a	2890.05 ± 14.45	2934.47 ± 14.68	2728.07 ± 13.64	4587.44 ± 22.94

K_E , gain of spectroscopy amplifier; MeV, megaelectron volt; ph, photon; phe, photoelectron; $PP_{1\text{phe}}$, peak position of a single photoelectron spectrum; PP_e , Compton edge peak position of a ^{137}Cs .

^a The values in this row are {Explanation}.

and λ is the wavelength in nanometers. The quantum efficiency was calculated as approximately 21% and the light output was estimated by dividing the quantum efficiency by the light yield.

The fabricated plastic scintillators exhibited a light output of approximately 59–64% of the output of the BC-408 scintillator (Saint-Gobain). To increase this value, naphthalene, an organic compound that is often used to improve solubility, could be added during the preparation process. In addition, other liquid monomers such as SR9035 and SR9036 could be used instead of styrene. Adding the aforementioned materials may improve scintillator properties.

4. Conclusion

Using a polymerization reaction in a high temperature heater, three plastic scintillators were fabricated to compare their properties with those of a commercial plastic scintillator. Styrene, PPO, and POPOP as the scintillator materials were mixed inside a 100-mL beaker and placed into a high temperature heater for approximately 250 hours. Surface machining operations such as cutting and polishing were performed and plastic scintillators of 4.5 cm diameter and 2.5 cm length were fabricated. Three standard gamma ray sources were used to evaluate the characteristics of the fabricated scintillators. Pulse spectra were measured via PMT, amplifier, high voltage power supply, and MCA using different gamma ray sources. Linearity and light output were calculated through pulse spectra analysis. Based on energy calibrations, the fabricated scintillators possessed more than 99.7% energy linearity. The scintillators showed a light output of approximately 59–64% of that of a BC-408 scintillator (Saint-Gobain). In the future, to improve the light output, other materials such as naphthalene and the new liquid monomers will be added and are expected to improve the properties of the scintillator. Studies related to fast neutron and charged particle detection will also be conducted to apply the fabricated scintillator in the field of radiation security.

Conflicts of interest

All authors have no conflicts of interest to declare.

Acknowledgments

This work was supported by a research grant of Hanyang University (Seoul, Korea; grant number, HY-2009).

REFERENCES

- [1] H. Penttilä, Characterization of a New Plastic Scintillation Material and Comparison with Liquid BC-501A Scintillator (Saint-Gobain, Paris, France), Oleksii Poleshchuk, Jyväskylä, Finland, 2015.
- [2] D. Reilly, N. Ensslin, H. Smith Jr., S. Kreiner, Passive nondestructive assay of nuclear materials, in: Doug Reilly, Norbert Ensslin, Hastings Smith Jr. (Eds.), Los Alamos National Lab, Los Alamos, NM, USA, 1991.
- [3] Z. Li, W. Chong, H. Yuekun, Z. Xiaojian, S. Feng, Z. Sun, W. Jinjie, A. Henghua, Z. Yuda, Z. Ziping, W. Yifang, Properties of plastic scintillators after irradiation, Nucl. Instrum. Methods A 552 (2005) 449–455.
- [4] M. Bertolaccini, C. Bussolati, S. Cova, I. De Lotto, E. Gatti, Optimum processing for amplitude distribution evaluation of a sequence of randomly spaced pulses, Nucl. Instrum. Methods Phys. Res. A61 (1968) 84–88.
- [5] M. Moszynski, M. Kapusta, M. Mayhugh, D. Wolski, S.O. Flyckt, Absolute light output of scintillators, IEEE Trans. Nucl. Sci. 44 (1997) 1052–1061.
- [6] Organic Scintillation Materials, Saint-Gobain Crystals, Paris, France.
- [7] G.H. Kim, C.H. Park, C.H. Jung, K.W. Lee, B.K. Seo, Development of the ZnS(Ag)/BC-408 phoswich detector for monitoring radioactive contamination inside pipes, J. Korean Assoc. Radiat. Prot. 31 (2006) 123–128.
- [8] B. Richard, Firestone Table of Isotopes, eighth ed., Wiley, New York, 1999.
- [9] Photomultiplier Tubes and Related Products, Hamamatsu Photonics Co., Hamamatsu City, Japan, 2010.